BNWL-1183 8
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QUARTERLY RESEARCH REPORT TO THE NASA MANNED SPACECRAFT CENTER

THE MEASUREMENT OF RADIATION EXPOSURE OF
ASTRONAUTS BY RADIOCHEMICAL TECHNIQUES

N72 - 13055
NASA- CR- 124713
Code   G3
Category 04
Pages 45

January 4, 1971 Through April 4, 1971

by

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April 15, 1971

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ABSTRACT

The concentrations of the radioisotopes observed in the feces from the Apollo 12 and 13 missions were normalized to the weight of the respective stable element in the specimens. These newly normalized concentrations confirmed all prior conclusions regarding assignment and identification of samples, anomalously high and low concentrations of radionuclides, and cosmic radiation dose.

The concentrations of 23 major, minor, and trace elements in the fecal samples from the Apollo 12 and 13 astronauts are reported. Most elemental excretion rates are comparable to rates reported for earlier missions. Exceptions are noted for calcium, iron, and tin. Body calcium and iron losses appear to be reduced during the Apollo 12 and 13 missions such that losses now seem to be insignificant. Refined measurements of tin excretion rates agree with normal dietary intakes. Earlier reported tin values are in error.

A new passive dosimetry canister has been designed which contains foils of tantalum, copper, titanium, iron, cobalt, aluminum, and scandium. This unit weighs only ~ 11 g more than the original design. By measuring the concentrations of the various products of nuclear reactions in these metals after space exposure, the characteristics of the incident cosmic particles can be determined.

A ^{210}Po concentration of $(2.58 \pm 0.41).10^{-4}$ d/m/cm^2 has been measured in a blank foil of the Solar Wind Composition experiment material exposed to the lunar atmosphere during the Apollo 12 landing. Analysis of the actual exposed foil is proceeding. A net increase in ^{210}Po activity, attributable to lunar exposure, can be correlated with the radon concentration of the lunar atmosphere. (At time of writing a real net activity attributable to lunar radon has been observed - the first such measurement of lunar atmosphere.)

The text of a paper entitled, "The Measurement of Radiation Exposure of Astronauts by Radiochemical Techniques" is included as an appendix.

TASK - DETERMINATION OF THE RADIONUCLIDE CONTENT OF FECES AND URINE
                FROM ASTRONAUTS ENGAGED IN SPACE FLIGHT

Astronauts engaged in space flight are subjected to cosmic radiation
which induces radioactive isotopes in their bodies. The radiation dose
received from cosmic particles can be determined from the quantities of
these induced radionuclides.[1] The concentrations of the induced radio-
activities can be determined by direct whole body counting of the astronaut
or by indirect measurement, such as counting that fraction of the radio-
nuclides excreted in the feces and urine. This latter approach was used
for evaluation of radiation activation during the course of the Apollo 12
and 13 missions. In addition, some fallout and naturally occurring radio-
isotopes have been measured, and variations in their concentrations may serve
as tracers of changes in the biological life processes occasioned by the
space environment.

The concentrations of the radioisotopes listed in Tables I and II have
been normalized by dividing the decay corrected disintegration rate by the
weight of the respective stable element in the sample determined by a tech-
nique of instrumental neutron activation analysis.[2-4] The specific
activities for urine specimens should be very nearly the same as those
present in the astronaut's body at the time of sampling since all elements
excreted in the urine must have been previously metabolized. The data for
feces is not quite so clear cut, however, since the quantities of inert
elements excreted can be perturbed by unmetabolized elements passing
through the gastrointestinal tract or by external addition, as is the case
with the sodium salt bactericide. This more precise method of normalizing
the data does not change any of the original conclusions [4, 5] regarding
assignment and identification of samples, anomalously high and low

concentrations of radionuclides, and cosmic radiation dose.

A paper entitled, "The Measurement of Radiation Exposure of Astronauts by Radiochemical Techniques," which is based on the measurements of the quantities of the cosmogenic radionuclides found in the feces and urine of the Apollo 7 through 13 astronauts, was presented on March 1, 1971 at the National Symposium on Natural and Manmade Radiations in Space. The text of this paper, which will be published in the proceedings of this symposium, is reproduced in Appendix A of this report.

TASK - NEUTRON ACTIVATION ANALYSIS OF FECES AND URINE FROM ASTRONAUTS ENGAGED IN SPACE FLIGHT

This program has been instituted in an attempt to foresee any possible metabolic changes in astronauts caused by conditions of weightlessness and prolonged physical inactivity which are manifested by an uptake or loss of an element or elements by their bodies. The primary concern is the terrestrially observed phenomenon of osteoporosis (loss of skeletal calcium), although changes in the uptake and excretion rates of other essential microconstituents of the body, such as cobalt, iron, selenium, and the alkali metals, are also important.

. A previously described technique of instrumental neutron activation analysis [1, 2, 4] was used to determine the concentrations of Ca, Na, K, Rb, Cs, Fe, Co, Zn, Cr, Sc, Br, Se, Hg, Ag, Sb, Au, Sn, As, Eu, Tb, Th, Hf, and Ta in the returned Apollo 12 and 13 fecal samples. These concentrations are reported in Tables III through VI.

Calcium and the Alkali Metals

The functional responsibility and biological importance of calcium and the alkali metals in the body is well known and has been discussed previously[4]. The fecal excretion rates of these elements are calculated from the data in Table III by dividing the total weight of each element by the number of man days of the mission.

Calcium fecal excretion rates of 0.302 and 0.39 g/man day for the Apollo 12 and 13 missions respectively are significantly lower than the rates observed for previous missions[4]. These low excretion rates indicate a negligible body calcium loss for these astronauts if they are ingesting a reasonable amount of calcium in their food (such as that ingested on the earlier manned Apollo missions).

Sodium fecal excretion rates of 58 and 78 mg/man day for the Apollo 12 and 13 missions seem to indicate that the specimens were not contaminated with a sodium salt bactericide. Assuming 2.76% of the excreted sodium is in the feces,[6] total loss rates of 2.1 and 2.8 g/man day are calculated which are similar to other determinations obtained for astronauts[4] and to normal dietary intakes[6].

The respective potassium fecal excretion rates are 250 and 304 mg/man day for the Apollo 12 and 13 astronauts. A potassium fecal excretion of 16.5%[6] leads to a total loss of 1.5 and 1.84 g/man day which is similar to intake and excretion values obtained for earlier missions[7].

Rubidium fecal excretion rates of 379 and 667 µg/man day become total body excretion rates of 1.65 and 2.90 mg/man day when divided by a 23%[8] fecal excretion. These values compare favorably with a normal daily intake of 2.53 mg[6, 9].

The cesium fecal excretion rates are 0.945 and 1.16 µg/man day for the Apollo 12 and 13 missions respectively. These are the lowest values yet observed for manned Apollo missions[3, 4].

Elemental Groups IB, IIB, VIA, and VIII

The physiological functions of the metals located in the center of the periodic table have been discussed in an earlier report[4]. While not all of these elements have known uses in the body, some have suspected essential properties and others are known to be toxic. The concentrations of the elements of this group which were measured are given in Table IV.

The chromium fecal excretion rates observed for Apollo 12 and 13 are 28.9 and 48.8 µg/man day respectively. These values are also the lowest yet observed[3, 4] and are considerably less than a normal intake of 150 µg/day[9]. This may be a reflection of dietary intake.

Iron fecal excretion rates for these latter two missions are 5.26 and 7.90 mg/man day. These values are in accord with the intake values for previous missions and indicate that the large loss of body iron observed for the Apollo 7 through 11 missions[7] has apparently been curtailed.

The measured cobalt fecal excretion rates are 4.79 and 6.25 μg/man day for the Apollo 12 and 13 missions. These values are much lower than rates observed for earlier missions[4] and are more nearly that which would be expected from normal dietary intake values[6, 9]. Intake values and urinary excretion rates should be checked to adequately evaluate the gain or loss of this element.

Silver fecal excretion rates of 17.7 and 12.6 μg/man day are calculated for the Apollo 12 and 13 missions and are the lowest values yet observed.

Respective gold losses are 165 and 21.7 μg/man day. The large difference between the two missions and the extremely high concentrations of gold in the three samples listed in Table IV as the Command Module Pilot, indicate the presence of this element in the astronauts' bodies is largely a very individual matter.

Zinc fecal excretion rates of 3.70 and 8.33 μg/man day for Apollo 12 and 13 astronauts respectively are the lowest values yet observed[2-4] although 8.33 μg/day is still within the expected range based on normal dietary intakes.

Elimination rates for mercury are calculated to be 13.7 and 21.3 μg/man day for these two missions which are comparable to normal dietary intakes. These mercury elimination rates are lower than previously observed rates [3, 4] by factors of 2-12.

Elemental Groups IV B, V B, VI B, and VII B

The body chemistry and/or toxic properties of these elements from the right side of the periodic chart has also been discussed elsewhere[4]. The concentrations and total weights of the measured elements in this category are given in Table V.

The tin fecal excretion rates are 1.41 and 16.2 mg/man day respectively for the Apollo 12 and 13 missions. These values are about three orders of magnitude higher than those reported for previous missions[3, 4] and are more compatible with normal dietary intakes of 17 to 22 mg/day[6, 9]. Discovery of a procedural error indicates that previously reported values of tin concentrations are incorrect. Corrected values will be given in a later report.

Arsenic has not been previously reported. However, the Apollo 12 and 13 mission fecal excretion rates are calculated to be <27 µg/man day and 2.85-13 µg/man day respectively. These values are considerably lower than the reported normal value of 2.3 mg/day[6].

Calculated antimony fecal excretion rates of 11.0 and 8.28 µg/man day for the Apollo 12 and 13 missions respectively are very similar to those observed on previous missions[3, 4]. One specimen (Apollo 12 CMP 225 hrs.) has an unprecedented high concentration of antimony.

Selenium fecal excretion rates of 14.7 and 20.4 µg/man day for the latter two missions are the lowest ever observed, but the significance of this fact is uncertain.

Fecal excretion of bromine, a minor path of elimination for this element, proceeded at the rate of 304 and 66.8 µg/man day for the Apollo 12 and 13 missions respectively. These rates are comparable to those previously observed[3, 4]. Three samples from the Apollo 12 mission have unusually high

bromine concentrations, and this circumstance may be sufficient to fingerprint the unlabeled specimen as coming from the LMP.

The Lanthanides, Actinides, and Groups III A, IV A, and V A

While the metabolic functions of these elements are not yet known, the data are reported in the hopes that they will prove useful in the future. Perhaps the concentrations and ratios of concentrations of these elements would be useful in the identification of samples. Of the elements reported in Table VI, only scandium has appeared previously. The fecal excretion rates calculated for the Apollo 12 and 13 missions for this element are 446 and 409 ng/man day, which are only slightly lower than previously observed values[4]. Normal daily intakes or excretion rates are not known for any of these reported elements. The measured fecal excretion rates are 136 and 128 ng europium/man day, (105-350) and (136-380) ng terbium/man day, 1.94 and (1.05-1.2) µg thorium/man day, 1.37 and 1.75 µg hafnium/man day, and 1.16 and 1.33 µg tantalum/man day.

Summary

The concentrations of 23 elements were measured in the fecal samples collected during the Apollo 12 and 13 missions. Most elemental excretion rates are comparable to those reported for earlier missions and with expected excretion rates based on normal dietary intakes where known. Major differences are observed in these latter two missions for calcium, iron, and tin. The excretion rate of calcium is lower than previously observed, and any body calcium loss is expected to be even less significant than it was for the earlier missions. The excretion of iron is greatly reduced in the Apollo 12 and 13 specimens to the point where loss of body iron by astronauts may no longer be a concern. The tin excretion rates reported for the latter missions

are about three orders of magnitude higher than reported for the earlier
missions. The higher values are more nearly those expected on the basis of
normal dietary intake, and the lower values reported earlier are felt to be
incorrect due to a procedural error. There is a possibility that not all
inflight fecal specimens from the Apollo 12 mission were returned since some
could have been jettisoned with the lunar excursion module. If this were
the case, the actual excretion rates for this mission would be higher than
those reported by an amount which would be dependent on the quantity of
specimens jettisoned. Due to the similarity of the reported excretion rates,
for this mission with other Apollo missions, it is likely that all samples
were indeed returned.

TASK - <u>INDUCED RADIONUCLIDES IN SPACECRAFT</u>

In order to more accurately determine the cosmic-ray flux and energy spectrum incident on astronauts during space flight, which is responsible for and relatable to the radiation dose received by them, an assembly of pure monitor foils has been developed. When these pure metals are exposed to the cosmic particle flux, various nuclear reactions will take place which are representative of the target element and the quantity and energy of incident particles. By measuring the concentrations of the various products and knowing the probability (cross section) for each observed reaction, the number and energy of incident particles can be calculated.

The assembly of metal foils was incorporated into the passive dosimetry cannister. A new cannister was designed with positive sealing screw caps and a thin (0.0075") side wall to reduce the attenuation of cosmic particles entering the can. The can was then lined with concentric sleeves of 0.003" tantalum, 0.001" copper, 0.001" titanium, 0.002" iron and 0.003" cobalt side by side as a single sleeve, and 0.004" aluminum foils. The standard dosimeter cluster, with the addition of about a 1/2" length of 0.020" diameter scandium wire sealed in glass, completes the cannister assembly. The entire unit only weighs ~1 g (7%) more than the original system. An exploded view of the new design is shown in Figure 1. The tantalum, copper, titanium, iron, and aluminum foils are intended as proton flux monitors, and the excitation functions for the production of many of the spallation products anticipated to be present in these foils after space exposure are well known. The cobalt foil and the scandium wire are intended as thermal and epithermal neutron monitors since the capture cross sections and decay characteristics of the products are ideally suited for measurement after extended space missions.

One of these new dosimeter assemblies should be flown on each of the
remaining Apollo missions to determine if any design changes, such as type
or quantity of metal foil, should be incorporated before use of this system
on project Skylab missions.

TASK - SEARCH FOR LUNAR ATMOSPHERE

Radon decay product analysis of the Solar Wind Composition (SWC) foils exposed to the lunar atmosphere by the Apollo 12 astronauts is presently in progress. The radon atoms present in the lunar atmosphere from the decay of surface uranium should embed themselves in the SWC foil and, through several rapid radioactive decays, be transformed into long-lived ^{210}Pb. By measuring the concentration of ^{210}Po (granddaughter of ^{210}Pb) in the returned SWC foils, it should be possible to characterize the radon concentration in the lunar atmosphere and, therefore, the uranium concentration in the lunar soil.

In order to obtain maximum sensitivity, a new low level Ortec 325S alpha counting system is being calibrated for the determination of ^{210}Po. The system has an absolute efficiency of 31.3% for this isotope and a background of 0.0002 counts per minute. The ^{210}Po in the SWC foils is separated by dissolution of the foil and autoplating onto a silver disc which is then counted. The silver disc and the reagents used in the process contribute a negligible amount to the measured ^{210}Po activity. Blank SWC foil G 30-11 (0.2118g, 53.24 cm^2) has been processed, and an activity of (2.58 ± 0.41) $.10^{-4}$ d/m/cm^2 was observed. This can be compared to a ^{210}Po activity of $(2.9-6.6).10^{-4}$ d/m/cm^2 observed previously in a similar blank foil measured or a less sensitive counting system. The Apollo 12 SWC foil exposed to the lunar atmosphere, G 17-7-6-7, will be analyzed for ^{210}Po content in near future.

EXPENDITURES

The following table documents the expenditures according to task and total cost incurred from January 4, 1971 through April 4, 1971 for the work reported herein.

TASK	EXPENDITURES
Determination of the Radionuclide Content of Feces and Urine From Astronauts Engaged in Space Flight	$ 4,062
Neutron Activation Analysis of Feces and Urine From Astronauts Engaged in Space Flight	4,062
Induced Radionuclides in Spacecraft	1,355
Search for Lunar Atmosphere	2,708
TOTAL COSTS	$12,187

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TABLE I

RADIOACTIVITY IN FECES FROM APOLLO 12 ASTRONAUTS*

Sample	dis/min ^{22}Na per g Na	dis/min ^{59}Fe per g Fe	dis/min ^{60}Co per g Co	dis/min ^{137}Cs per g Cs
Unlabeled				$(2.8 \pm 1.9).10^6$
LMP #2			$(6.1 \pm 2.4).10^4$	
LMP 23N				$(1.7 \pm 0.5).10^6$
CDR	56 ± 31		$(3.8 \pm 2.3).10^4$	
CMP GET 79		2100 ± 1000		
CMP 225 Hrs			$(2.5 \pm 1.5).10^4$	

* The radioactivities have been normalized by dividing the disintegration rate by the weight of the stable element and decay correcting to the splashdown date, 11-24-69.

TABLE II

RADIOACTIVITY IN FECES FROM APOLLO 13 ASTRONAUTS*

Sample	dis/min ^{22}Na per g Na	dis/min ^{59}Fe per g Fe	dis/min ^{60}Co per g Co	dis/min ^{137}Cs per g Cs
#1	5.6 ± 4.3	$(2.58 \pm 0.05).10^4$	$(7.4 \pm 3.0).10^4$	$(1.12 \pm 0.01).10^8$
#2	39 ± 17	$(3.00 \pm 0.07).10^4$	$(8.3 \pm 3.1).10^4$	$(1.75 \pm 0.02).10^8$
#3			$(2.2 \pm 0.9).10^5$	
#4	5.6 ± 1.8	$(2.78 \pm 0.05).10^4$	$(8.2 \pm 2.5).10^4$	$(6.63 \pm 0.13).10^7$
#5		$(5.46 \pm 0.83).10^3$	$(5 \pm 3).10^4$	$(8.8 \pm 1.8).10^6$
#6	0.99 ± 0.73		$(3 \pm 2).10^4$	

* The radioactivities have been normalized by dividing the disintegration rate by the weight of the stable element and decay correcting to the splashdown date, 4-17-70.

TABLE III

Concentrations of ... in ... Astronaut Fecal Samples

	Ca		Na		K		Rb		Cs	
	ppm*	g**	ppm*	mg**	ppm*	g**	ppm*	mg**	ppm*	µg**
Apollo 12										
LMP #1	?;?30	3.728	260	20.7	3,900	0.311	12.4	0.991	0.0554	4.41
Unshielded	8,970	1.86	2,780	609	10,000	2.2	6.63	1.45	0.0151	3.30
LMP #2	8,620	1.23	570	82	3,160	0.453	8.14	1.16	0.0219	3.13
CDP 23N	9,070	0.411	4,330	180	8,240	0.343	10.9	0.455	0.0508	2.11
CDR	?,??	0.912	95.8	12.7	4,090	0.544	12.3	1.63	0.0243	3.23
CMP GET 79	8,320	1.38	1,790	297	7,210	1.20	15.8	2.61	0.0287	4.76
CMP GET 101	7,390	0.808	2,640	288	7,580	0.829	8.89	0.972	0.0190	2.08
CDP 225 HRS	11,100	1.82	1,770	289	10,500	1.73	14.2	2.33	0.0358	5.87
Apollo 13										
#1	4,720	1.61	628	104	5,980	0.991	14.4	2.39	0.0246	4.07
#2	3,500	0.37	160	17	3,920	0.424	11.5	1.24	0.0204	2.21
#3	9,600	0.406	714	25.0	2,910	0.102	12.9	0.451	0.0257	0.900
#4	11,300	3.04	1,760	475	7,030	1.89	13.2	3.57	0.0251	6.77
#5	3,580	0.535	1,310	196	6,790	1.02	11.0	1.65	0.0157	2.35
#6	6,810	1.06	3,720	577	6,420	0.996	16.9	2.61	0.0281	4.36

* Net weight basis
** Total weight per defecation

TABLE IV

...TIONS IN ASTRONAUT FECAL SAMPLES

	Cr		Fe		Co		Ag		Au		Zn		Hg	
	ppm*	µg**	ppm*	mg**	ppm*	µg**	ppm*	µg**	ppm*	µg**	ppm*	mg**	ppm*	µg**
Apollo 12														
#1	1.69	176	162	12.9	0.131	10.4	0.419	33.4	0.341	27.2	126	10.1	0.531	50.3
	.09	125	97.2	21.3	0.0730	16.0	0.542	119	0.587	129	76.6	16.8	0.547	120
LMP #2	.19	171	207	29.6	0.165	23.6	0.404	57.8	0.255	36.5	131	18.7	0.840	120
LMP 23N	1.29	53.5	146	6.08	0.0905	3.77	0.376	15.7	0.353	14.7	115	4.78	0.607	25.3
CDR	0.256	88.6	167	22.3	0.160	21.3	0.204	27.1	0.00977	1.30	114	15.2	0.392	52.1
LMP GET 79	0.555	91.9	143	23.7	0.151	25.0	0.418	69.2	2.26	374	93.7	15.5	0.105	17.3
	0..0	90.1	105	19.5	0.124	13.5	0.480	52.5	3.40	372	75.2	8.22	0.0605	6.62
	1.07	175	204	23.5	0.200	32.8	1.01	165	4.45	729	146	23.9	0.160	26.2
Apollo 13														
#1	0.704	132	186	30.9	0.124	20.6	0.338	56.1	0.441	73.1	188	31.2	0.381	63.1
#2	1.30	173	128	14.9	0.114	12.3	0.137	14.9	0.00423	0.458	151	16.3	0.281	30.4
#3	0.384	13.4	207	7.24	0.0967	3.39	0.141	4.94	0.00212	0.0743	251	8.78	0.357	12.5
#4	0.777	209	180	48.4	0.158	42.6	0.258	69.4	0.00679	1.83	183	49.3	0.575	155
#5	0.957	14.	100	15.0	0.0881	13.2	0.268	40.1	0.352	52.7	126	18.8	0.236	35.3
#6	1.29	201	159	24.7	0.126	19.6	0.257	39.9	0.00540	0.838	158	24.4	0.546	84.7

* Wet weight basis

** Total weight per defecation

TABLE V

... Co and As ... ... FECAL SAMPLES

	Sn		As		Sb		Se		Br	
	ppm*	µg**	ppm*	µg**	ppm*	µg**	ppm*	µg**	ppm*	µg**
Apollo 12										
#1	5.30	0.303	<0.34	<27	0.320	25.5	0.509	40.5	1.98	158
...	3.50	1.75	<1.1	<20	0.153	33.5	0.298	35.3	26.4	5,790
#2	23.3	3.34	<1.6	<230	0.108	15.5	0.518	74.2	15.3	2,180
...	4.05	0.108	<2.2	<93	0.272	11.3	0.412	17.1	14.4	600
CDR	19.5	2.59	<0.21	<28	0.283	37.6	0.479	63.7	0.843	112
... 79	26.2	4.34	<0.41	<69	0.158	26.1	0.380	62.9	1.27	210
...	7.34	3.731	<0.37	<40	0.217	23.7	0.315	34.4	1.19	130
... 225 HRS	5.77	1.11	<0.56	<92	0.299	164	0.561	91.9	0.621	102
Apollo 13										
#1	162	26.9	<0.19	<31	0.148	24.5	0.465	77.1	1.07	177
#2	2.98	0.312	0.157	17.0	0.124	13.5	0.383	41.5	0.573	62.1
#3	322	11.3	<0.27	<9.4	0.125	4.36	0.620	21.7	2.27	79.6
#4	173	46.6	<0.34	<91	0.170	45.8	0.426	115	1.68	453
#5	13.3	1.99	<0.23	<35	0.159	23.7	0.289	43.3	0.989	148
#6	59.1	9.17	<0.33	<52	0.233	36.1	0.420	65.1	1.76	274

* Wet weight basis

** Total weight per defecation

TABLE VI

...IS IN ASTRONAUT FECAL SAMPLES

	Eu		Tb		Th		Sc		Hf		Ta	
	ppm*	µg	ppm	µg	ppm	µg	ppm*	µg	ppm	µg	ppm*	µg**
Apollo 12												
	.0236	98	.0036	< 350	0.0374	2.98	0.0268	2.14	0.0247	1.97	0.0164	1.31
	0.00110	542	0.0017	< 360	0.0151	3.30	0.00921	2.02	0.0100	2.19	0.00712	1.56
	...	132	.0029	< 520	0.0155	2.22	0.00766	1.10	0.00725	1.04	0.00757	1.08
	...	--	...	110	...	1.47	0.0270	1.12	0.0165	0.685	0.0164	0.681
	...	119	0.0033	< 240	0.0189	2.51	0.0105	1.39	0.0162	2.16	0.0191	2.54
	0.000111	104	< 0.0024	< 300	0.0169	2.31	0.0107	1.77	0.00993	1.65	0.00856	1.42
	0.000056	104	< 0.0023	< 250	0.00958	1.05	0.00970	1.06	0.0109	1.19	0.0119	1.30
	0.00167	273	0.00651	1,070	0.0208	3.41	0.0185	3.03	0.0185	3.03	0.0119	1.94
Apollo 13												
#1	0.000676	165	0.0021	210	0.00340	0.57	0.00976	1.45	0.00918	1.52	0.00743	1.23
#2	0.000720	78.0	< 0.0019	< 200	0.00461	0.499	0.00694	0.752	0.00706	0.764	0.00708	0.767
#3	0.000519	18.2	< 0.0021	< 74	0.00509	0.176	0.00553	0.193	0.00394	0.138	0.00247	0.0865
#4	...	--	... 0.0001	110	0.0103	2.91	0.00280	2.64	0.0188	5.06	0.0107	2.88
#5	0.000610	91.3	< 0.0021	< 310	0.00326	0.488	0.00684	1.02	0.00828	1.24	0.00542	0.811
#6	0.00116	179	< 0.0034	< 520	0.0139	2.15	0.00807	1.25	0.0108	1.68	0.0138	2.14

* Wet weight basis

** Total weight per defecation
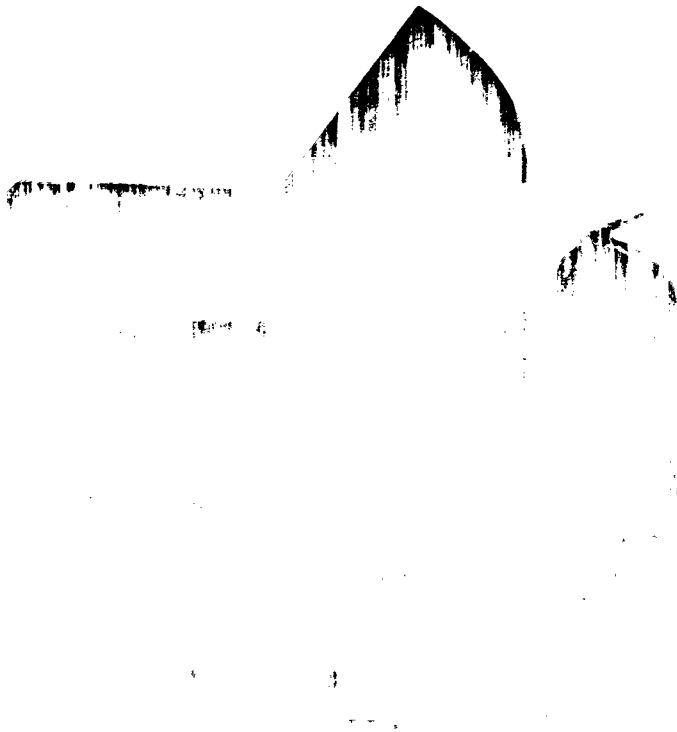
FIGURE 1.

Exploded view of the individual passive dosimetry container. From left to right and top to bottom: 0.003" tantalum foil; 0.001" copper foil; 0.002" iron foil; 0.004" aluminum foil; 0.003" cobalt strip; 0.001" titanium foil; resonance measure; dosimeter cluster including 0.020" scandium wire sealed in glass at top center; aluminum can; teflon "0" ring seal; screw end cap with swivel mounting bracket.

APPENDIX A

THE MEASUREMENT OF RADIATION EXPOSURE OF
ASTRONAUTS BY RADIOCHEMICAL TECHNIQUES

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ABSTRACT

The principal gamma-ray-emitting radioisotopes produced in the body of astronauts by cosmic-ray bombardment which have half-lives long enough to be useful for radiation dose evaluation are ^7Be, ^{22}Na, and ^{24}Na. The sodium isotopes were measured in the preflight and postflight urine and feces, and those feces specimen collected during the manned Apollo missions, by analysis of the urine salts and the raw feces in large crystal multidimensional gamma-ray spectrometers. The ^7Be was chemically separated, and its concentration measured in an all NaI(Tl), anticoincidence shielded, scintillation well crystal.

The overall sensitivity of the experiment was reduced by almost all variables such as low concentrations of excreted cosmogenic radionuclides, high concentrations of injected radionuclides, low sample sizes, long delay periods before analysis, and uncertain excretion rates. The astronaut radiation dose in millirads, as determined by this technique, for the Apollo 7, 8, 9, 10, 11, 12, and 13 missions was 320, 160, <3'5, 870 $\pm$ 550, 31, 110, and <2 respectively. In view of these limitations this technique would be best applied to cases of unusually high exposures, such as that encountered from solar flares.

INTRODUCTION

With the advent of space flight, it has become necessary to determine the radiation dose to man from exposure to the galactic, Van Allen, and solar flare particles. The high-energy galactic portion of the spectrum is fairly constant and has a relatively low intensity. The high intensity Van Allen radiation is of medium energy and localized in space. However, the solar radiation is not so predictable, and the flux and energy of particles from the sun can vary tremendously depending on solar activity. Since high levels of radiation exposure are possible, radiation dosimetry which will properly define radiation exposures is essential in space research programs. Dosimetry methods employed thus far, such as nuclear emulsion films, thermoluminescent dosimeters, and ionization gauges provide very useful indirect methods for estimating radiation dose but are subject to limitations. They measure only a surface exposure at a specific point(s) in the spacecraft or on the astronaut's body rather than an integral whole body exposure, and they have a limited sensitivity to large variations in particle energy. Some of the inherent limitations of these external dosimeters are avoided by using the induced radioactivity in the body of an astronaut as a measure of his radiation exposure. During a space flight, radionuclides are produced throughout the entire body of an astronaut, and the production rates are related directly to the cosmic particle flux within the body. The absolute and relative amounts of the various radionuclides bear a direct relationship to the intensity and energy spectrum of the particles which are doing the biological damage.

The radiation dose received from the cosmic particles can be determined from the quantities of induced radionuclides[1,4]. The amounts of these induced activities are determined by direct measurement, i.e., whole body counting of the radioactivity ( ), or by other means such as counting the

radionuclides excreted in the feces and urine. The latter approach was used for evaluation of radiation activation during the course of the manned Apollo missions.

The principal gamma-ray-emitting radioisotopes produced in the body by cosmic-ray bombardment are ^7Be ($t_{1/2}$ = 53 day), ^{11}C ($t_{1/2}$ = 20.5 min), ^{13}N ($t_{1/2}$ = 9.96 min), ^{22}Na ($t_{1/2}$ = 2.60 yr), and ^{24}Na ($t_{1/2}$ = 15.0 hr). The primary mode of production of ^7Be and ^{11}C is the spallation of carbon, nitrogen, and oxygen in the body. The ^{13}N comes principally from the spallation of nitrogen and oxygen, the ^{22}Na from the spallation of sodium, phosphorus, and calcium, and the ^{24}Na from the neutron activation of natural sodium. Of these, ^{11}C and ^{13}N are too short-lived to be measured by any method other than a direct determination, and this direct counting would have to be done as soon as possible after recovery. This is unfortunate, since these radioisotopes are produced in the largest abundance. The radionuclides ^7Be, ^{22}Na, and ^{24}Na are, however, sufficiently long-lived to facilitate their use in making dose estimates from measurement of their quantities in urine and fecal samples.

Other radioisotopes were also expected to be present in the bioassay samples. In addition to the aforementioned cosmogenic radionuclides, measurements of naturally present ^{40}K, normally occurring ^7Be, ^{22}Na, and ^{137}Cs, and ^{51}Cr and ^{59}Fe which were injected for medical studies were also made. Another radioisotope, ^{60}Co, was detected and quantitatively measured in some of the specimens. Corrections to the cosmogenic ^7Be and ^{22}Na must be made to account for the quantities of these radioisotopes normally occurring in the body because of fallout, food intake, and other ingestion processes. The quantities of the naturally occurring ^{40}K and the injected ^{51}Cr and ^{59}Fe in the bioassay samples could serve as biological markers of certain changes of metabolic processes during the course of a mission.

In previous studies, induced radioactivity to radiation dose relation-
ships have been established for the radionuclides ^7Be, ^{22}Na, and ^{24}Na as a
function of energy for proton bombardment of muscle tissue[2]. From these
relationships and from the ratios in which these radionuclides are produced,
the "effective proton energy" of cosmic radiation incident on an astronaut can
be determined. This allows the direct estimation of the whole body radiation
exposure received by astronauts from measurements of the radionuclides produced
in their bodies.

EXPERIMENTAL

Preflight and postflight urine and feces and those feces specimens
collected in flight were analyzed. Due to the quarantine period following
lunar landing missions, all samples were not immediately available for analyses,
thus allowing the short-lived radionuclides to decay. The urine specimens
which were of small volume were solidified prior to analysis by the addition
of $CaSO_4$ to 25 ml or less of the raw urine in order to form a standard counting
geometry. Any samples of initial volume greater than 25 ml were treated by
repeatedly boiling to dryness with nitric acid to destroy the organic matter
present. The remaining salts were counted in large crystal multidimensional
gamma-ray spectrometers[-] for determination of ^{22}Na, ^{24}Na, ^{40}K, ^{51}Cr, ^{59}Fe,
^{60}Co, and ^{137}Cs. The salts were then dissolved in a weak HCl solution and
diluted to known volume. An aliquot of this solution was taken for neutron
activation analysis to determine the concentrations of stable elements in the
sample. The remainder of the solution was reduced in volume to approximately
5 ml and transferred to a 100 ml polypropylene centrifuge tube. Approximately
mg of the carrier and 25 mg of the carrier were added, and the solution
was neutralized with concentrated K_4OH. After centrifugation the supernatant
solution was discarded. Thirty-five ml of HCl was added to the remaining
tube. After the solution was stirred vigorously until the salts dissolved. Upon centrifugation the

supernatant liquid was transferred to a clear centrifuge tube, saturated with
NH_4Cl, and heated in a water bath. If necessary, additional NH_4Cl was added
until a $Be(OH)_2$ precipitate settled from the solution. The solution was
then centrifuged, and the supernatant fraction was discarded. The resulting
quantitative precipitate containing the ^7Be activity was counted in an all
NaI(Tl) anticoincidence shielded, 7-inch diameter scintillation well crystal
in the absence of all interfering activities. This was necessary in order
to measure the relatively small quantities of ^7Be present.

Fecal samples were thoroughly mixed in their collection bags to ensure
homogeneity of the specimens. A small corner was cut off each bag and
aliquots were extruded into standard counting geometry containers for measure-
ments on multidimensional gamma-ray spectrometers   of   the   radioisotopes
^{24}Na, ^{40}K, ^{51}Cr, ^{59}Fe, ^{60}Co, and ^{137}Cs. Separate aliquots were wet ashed with
nitric acid and hydrogen peroxide to destroy the organic matter present.
The resulting salts were dissolved in dilute nitric acid, and the same pro-
cedure as above was followed for separation of the ^7Be activity.

A luminous material composed of ^{147}Pm microspheres mixed with a scintillator
is used extensively in the spacecraft in acrylic switch tips and sighting
figures used in docking maneuvers. Because of the high rejection rate of
within this caused by pinhole leaks, there is some concern about the
possible presence of ^{147}Pm in the weightless space capsule environment. For
this latter mission, approximately 10 mg of mixed rare earths were added
to these prior to wet ashing. These materials serve as carriers for ^{147}Pm,
which could possibly have been ingested in the crew members. This rare earth
fraction was separated from the beryllium fraction after the initial NH_4OH
precipitation by dissolving the precipitate in approximately 8 ml of 3M HCl
at a pH of of 40 mixes 5 40. Centrifugation separates the rare earth

precipitate from the beryllium in the supernatant solution. The rare
earth fraction was then dissolved in two parts concentrated HNO_3 and three
parts saturated boric acid solution and reprecipitated with NH_4OH. After
centrifugation and decantation, the precipitate was dissolved in dilute HCl;
and saturated oxalic acid solution was added to precipitate the rare earth
oxalates. The solution was centrifuged; the supernatant solution was
decanted; and the quantitative precipitate was washed with alcohol, trans-
ferred to a 1-inch diameter stainless steel dish and counted in an end
window, gas flow beta counter for the measurement of ^{147}Pm.

RESULTS

The results of the individual determinations are given in Tables I
through IV. All data have been normalized to a gram of feces, a milliliter
of urine, or a gram of the respective stable element as determined by a
technique of instrumental neutron activation analysis[8]. All data have
been decay corrected to the time of splashdown of each respective mission.
The results of all the radionuclide measurements in the excreta are given in
the tables although only the concentrations of the cosmogenic radionuclides
^{7}Be, ^{22}Na, and ^{24}Na are of importance for the subject matter of this communica-
tion. The various samples in the tables are listed by the letters A, B, and
C or CM, CP, and CDR to identify an individual astronaut. Those samples
listed by numbers are unidentified and arbitrarily coded. The collection
time for each specimen is given as Pre-, In-, or Post-flight unless more
detail is known, in which case a number refers to elapsed time into the
mission in hours, the letter F followed by a number indicates that number
of days prior to flight. F+0 refers to the first voiding after splashdown,
this is the first 24 hour collection after splashdown and Day 2 is the
following day after splashdown.

The average values of the cosmogenic radionuclide concentrations in each basic flight period are summarized in Table V according to the various methods of normalization. The increase in the activities from preflight to inflight and postflight periods should be indicative of the exposure to cosmic radiation. The concentrations of each radionuclide increase rather regularly for the Apollo 7 mission regardless of the method of normalization. However, the fecal data for the Apollo 8 mission are quite irregular, with only the urine data demonstrating increases in the cosmogenic radionuclides. The Apollo 9 and 13 missions show increases in the ^7Be concentration in the urine but demonstrate decreases in the ^{22}Na concentrations while the reverse is true for Apollo 11. Regular increases are shown for Apollo 10 and 12.

The increases in cosmogenic radioactivity from preflight levels to those after exposure to the space environment are almost certainly due to cosmic particle activation. Equating the magnitude of the increase with the radiation dose delivered by the particles is still fairly difficult, particularly when the dose is quite small as has been the case on all manned Apollo missions thus far. Concentrations normalized to the unit mass or volume of excreta are subject to variation in the biological dilution of the specimen. Concentrations normalized to the unit mass of stable element in the feces are subject to variation in the quantities of unstabilized elements passing through the gastrointestinal tract. Only the quantities of radionuclides in urine normalized to the amount of stable element present can be expected to be reasonably representative of the specific activity in the whole body since the urine contains only metabolized material.

In order, it is necessary to make some assumptions regarding the percentages of a given stable element excreted in the feces or urine, the average daily output of feces and urine, and the "contamination" of feces by urine to reconcile the specimen data in order to correlate the data with the experimental

results for proton irradiated muscle tissue[1,2], proton irradiated radio-
therapy patients[3], and neutron irradiated radiotherapy patients[4]. In this
manner, the average effective proton energy incident on the astronauts and
the radiation dose received by them can be estimated. The details of these
calculations will be omitted here since they are given elsewhere[9-12]. The
results indicate an average effective proton energy of 38-40 MeV incident on
the Apollo 7 mission astronauts and <38 MeV on the Apollo 8 mission astro-
nauts. Radiation doses of 480 + 310, <375, 870 + 550, <480, and <250 millirads
for the Apollo 7, 9, 10, 12, and 13 missions respectively are calculated.

Since the specific activity of the cosmogenic radionuclides in the urine
should be a more accurate representation of the whole body burden of induced
radioactivity, the specific activity of the ^{22}Na in the postflight urine of
astronauts is compared to the specific activity of ^{22}Na in the urine of
radiotherapy patients who have received a known radiation dose. This com-
parison leads to estimated cosmic radiation doses received by the astronauts
on the Apollo 7, 8, 11, and 12 missions of 230, 160, 27, and 110 millirads
respectively. It should be pointed out here that the uncertainty of the
data given in Table 9, and hence of these results, is quite large in some
instances.

DISCUSSION

In principle the relationships between induced activity and radiation
dose are straightforward. The probability for production of a certain
isotope in the body of an astronaut is basically a function of the energy
of the proton. Similarly, the radiation dose from a cosmic proton is also
a function of its energy, and therefore the induced activity is logically
related to the radiation dose. Such relationships have been empirically
determined for several different situations, and it remains only to
determine the production rate for a particle irradiated
at a specific energy before the doses can be predicted.

In practice, however, the procedure is not quite as simple as that just described. A calibrated whole-body counter is required to determine the quantities of induced radionuclides, and a high sensitivity-low background instrument would be required to measure the small quantities of radionuclides induced by the low levels of cosmic radiation encountered on a normal space flight. In lieu of the availability of a suitable whole-body counter, an indirect approach such as that used in this work can be applied. The principal limitations to this method have already been touched upon above. Only a small and uncertain fraction of the induced activity is eliminated in the excreta. Thus only the specific activity of an induced radioisotope in the urine can be extrapolated to the whole body burden with a reasonable degree of accuracy.

While the efficiency of low-level sample counters is routinely several orders of magnitude higher than whole-body counters, the small fraction of the total body activity in any bioassay sample reduces the sensitivity of a specimen measurement to the point where it is little better than that of a whole-body count. To complicate the situation in this work even further, there is a large demand for aliquots of post-flight urine specimens from the astronauts and frequently only a small mass of a 24-hour collection has been available for radionuclide concentration measurements. An additional complication is the collection of non-isotope diluting classes (Apollo 13 excepted) in the collection of isotope from the astronauts for medical studies.

If these factors were not present the cosmogenic radionuclides in a sample could be determined to improve the overall accuracy of the experiment. In summary, the small mass of the lunar landing experiment, the small aliquots of the specimens which allowed only minimum counting statistics, these factors all contribute

to the reduced accuracy and sensitivity of the measurements reported herein. In an effort to improve the situation, a high sensitivity combination whole-body counter and sample counter has been proposed which could be rapidly utilized after a mission (even onboard the recovery vessel) to make accurate measurements of the whole body burden of radionuclides in the astronauts. The combination of direct measurement of whole body burdens of radionuclides and the early measurement of relatively large quantities of excreta should make much more accurate dose estimates possible.

This technique for measurement of radiation dose should be perfected during routine space missions so that in the event of an unusually high exposure, such as might be expected from a solar flare, an accurate deter-mination of the radiation dose can be obtained. This situation would be one apropos to those nuclear criticality accidents[3-15] where conventional dosimetry techniques were extended and clinical radioactivity was measured to interpret the radiation dose received by the exposed individuals.

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TABLE I

RADIONUCLIDES IN FECES FROM APOLLO ASTRONAUTS

ACTIVITY IN DISINTEGRATIONS/MINUTE/GRAM FECES ON DAY OF SPLASHDOWN

MISSION	SAMPLE IDENTIFICATION		^{7}Be	^{22}Na		^{51}Cr	^{59}Fe	^{60}Co	^{137}Cs	^{147}Pm
									0.167±0.028	
									0.178±0.090	
									0.147±0.025	
				0.003±0.1			0.102±0.06		0.19±0.02	
				0.005±0.1					0.07 0.02	
									0.15 0.04	
									0.22 0.09	
							0.59±0.06		0.13 0.03	
				0.30±0.			0.05 0.0		0.3 0.9	
				1.005±0.			0.0		0.0 0.9	
								0.57±0.003	0.07 0.06	
				0.02±0.					0.0 0.07	
								0.013±0.008	0.0 0.03	
									0.0 0.03	
				0.07±0.				0.005±0.0015	0.	
				0.02±0					0.0 0.03	
				0.0 0.0				0.073±0.016		
								0.003±0.004	0.	<147
									0.	
								0.07±0.031		
								0.07±0.006	0.	<115
								0.07±0.007	0.	<77.4
								0.0 0.013	0.	<73.2
			0.5					0.0 0.014		<24.9
				0.005±1						
				0.01±0						
								0.0 0.012		<0.698
								0.0 0.003		<0.057
										<0.04
										<0.5
										<0.5
									0.0	<5.5
							0.0±0.000			<5.0
									0.0	<5.07
				.00±0			0.0 0.057			<5.7
										<5.7
									0.122	

TAB E II

RADIONUCLIDES IN FECES FROM APOLLO ASTRONAUTS

MISSION	SAMPLE IDENTIFICATION	FLIGHT PERIOD	^{22}Na g Na	^{51}Cr/g Cr	^{55}Fe/g Fe	^{60}Co/g Co	^{137}Cs/g Cs
7	B	PRE		$(4.298 \pm 0.05) \cdot 10^{?}$	2 ± 150		$(8.3 \pm 1.4) \cdot 10^{6}$
..	C	PRE		$(4.32 \pm 0.10) \cdot 1^{?}$			
?	S/N 2276	IU		$(3.13? \pm 0.0?) \cdot 10^{?}$	1.10 ± 220		$(7.8 \pm 1.3) \cdot 10^{6}$
..	S/N 2277	I?'		$(1.6? \pm 0.0?) \cdot 10^{?}$			$(6.4 \pm 1.0) \cdot 10^{6}$
..	S/N 2278	I''	0.04 ± 1.47	$(2.1?? \pm 0.0?) \cdot$	70 ± 120		$(5.2 \pm 1.2) \cdot 10^{6}$
..	S/N 2282	.	$1.2? \pm 17$	$(2.6? \pm 0.0?) \cdot$	$24 \pm ??$		$(5.6 \pm 2.0) \cdot 10^{6}$
.				$(1.5? \pm 0.0?) \cdot$			$(5.59 \pm 0.90) \cdot 10^{6}$
.		.		$(2.7? \pm 0.0?) \cdot$			$(1.6 \pm 1.1) \cdot 10^{6}$
.		..		$(2.4? \pm 0.0?) \cdot$	$3.7? \pm 2?$		$(2.4? \pm 0.20) \cdot 10^{7}$
.		.	$0.5? \pm 26$	$(1.5? \pm 0.0?) \cdot$	$2 \pm 2?$		$(1.40 \pm 0.14) \cdot 10^{7}$
.		..		$(1.7? \pm 0.0?) \cdot$	$? \pm 2?$		$(3.6 \pm 1.2) \cdot 10^{6}$
.		POST	$5.0 \cdot .0$	$(1.1? \pm 0.1?) \cdot$	$2 \pm ?$		
.	:	POST		$(4.2? \pm 0.1?) \cdot 10^{4}$	$5.6? \cdot 1? \cdot 10^{4}$	$(1.956 \pm 0.068) \cdot 10^{6}$	$(1.461 \pm 0.096) \cdot 10^{7}$
.		.		$(1.0? \pm 0.1?) \cdot$	$? \pm ?$		$(4.58 \pm 0.48) \cdot 10^{6}$
..		..		$(1.7? \pm 0.?) \cdot$	$? \pm ?$		$(9.4? \pm 0.73) \cdot 10^{6}$
.		.	$0.7? \pm 16$	$(1.2? \pm 0.?) \cdot$	$? \pm ?$		$(3.3? \pm 0.74) \cdot 10^{6}$
		E		$(2.? \pm ?) \cdot$	$? \pm ?$	$(6.8 \pm 1.0) \cdot 10^{5}$	
		:		$(2.7 \pm 1.1?)$		$(6.? \pm 1.7) \cdot 10^{4}$	$(9.51 \pm 1.70) \cdot 10^{6}$
		.		$(2.? \pm 1.?)$	$? \pm :$		$(1.0? \pm 1.1?) \cdot 10^{7}$
				$(2.? \pm 0.?)$	$? \pm ?$		$(2.? \pm ?) \cdot 10^{7}$
						$(6.0? \pm 2.2) \cdot 10^{5}$	
	..	...		$(2.? \pm 1.?)$		$(? \pm 2.0) \cdot 10^{4}$	$(3.0? \pm ?) \cdot 10^{6}$
	.	.		$(2.? \pm 2?)$		$(? \pm 5.?) \cdot 10^{4}$	$(1.0? \pm 1.7?) \cdot 10^{6}$
	.	.		$(2.? \pm .?)$	.	$(? \pm 1.0?) \cdot 10^{4}$	$(? \cdot ?) \cdot 10^{6}$
	.			$(2.? \pm ..?)$		$(? \pm 2.0?) \cdot$	$(1.0? \pm .1?) \cdot 10^{6}$
:	...	...					
.		..					
.							

Activity in disintegrations/minute/gram inert element on day of splashdown

TABLE III

RADIOACTIVITY IN BODY TISSUES ... AND ASTRONAUTS

			Y	DIS	...	...	... DAY OF SPLASHDOWN		
TICKET		F				^{22}Na	^{40}K	^{60}Co	^{137}Cs
								0.00501±0.00042	0.250±0.002
									0.0096±0.0047
									0.134±0.1
						2.79±0.03	2.79±0.03		0.44±0.05
						1.00±2.1	1.00±0.02		2.1±0.17
							0.0097±0.022		2.1±0.1
							0.27±0.07		1.1±0.1
							0.27±0.03		2.1±0.2
									0.1
								0.0072±0.0027	
								0.0072±0.0010	

0.0072±0.013

$\cdot 10^6$

$\cdot 10^7$

$\cdot 10^6$